

# REPORT DOCUMENTATION PAGE

Form Approved  
OMB No. 074-0188

Public reporting burden for this collection of information is estimated to average 1 hour per response, including the time for reviewing instructions, searching existing data sources, gathering and maintaining the data needed, and completing and reviewing this collection of information. Send comments regarding this burden estimate or any other aspect of this collection of information, including suggestions for reducing this burden to Washington Headquarters Services, Directorate for Information Operations and Reports, 1215 Jefferson Davis Highway, Suite 1204, Arlington, VA 22202-4302, and to the Office of Management and Budget, Paperwork Reduction Project (0704-0188), Washington, DC 20503.

<b>1. AGENCY USE ONLY (Leave blank)</b>		<b>2. REPORT DATE</b> 1996	<b>3. REPORT TYPE AND DATES COVERED</b> Presentation, 23-25 July, 1996	
<b>4. TITLE AND SUBTITLE</b>  The Use of Carbon Aerogel Electrodes for Deionizing Water and Treating Aqueous Process Wastes			<b>5. FUNDING NUMBERS</b> DOE Contract No. W-7405-Eng-48.	
<b>6. AUTHOR(S)</b> J.C. Farmer, G.V. Mack, & D.V. Fix				
<b>7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES)</b>  Lawrence Livermore National Laboratory Livermore, CA 94550			<b>8. PERFORMING ORGANIZATION REPORT NUMBER</b>  N/A	
<b>9. SPONSORING / MONITORING AGENCY NAME(S) AND ADDRESS(ES)</b>  SERDP 901 North Stuart St. Suite 303 Arlington, VA 22203			<b>10. SPONSORING / MONITORING AGENCY REPORT NUMBER</b>  N/A	
<b>11. SUPPLEMENTARY NOTES</b> Presented at the Fourth International Congress on Environmentally Conscious Design and Manufacturing, Cleveland, OH, 23-25 July, 1996. This work was supported in part by DOE under Contract No. W-7405-Eng-48. The United States Government has a royalty-free license throughout the world in all copyrightable material contained herein. All other rights are reserved by the copyright owner.				
<b>12a. DISTRIBUTION / AVAILABILITY STATEMENT</b>  Approved for public release: distribution is unlimited			<b>12b. DISTRIBUTION CODE</b> A	
<b>13. ABSTRACT (Maximum 200 Words)</b>  A wide variety of ionic contaminants can be removed from aqueous solutions by electrosorption on carbon aerogel electrodes. Carbon aerogel is an ideal electrode material because of its low electrical resistivity, high specific area, and controllable pore size distribution. This approach may avoid the generation of a substantial amount of secondary waste associated with ion exchange processing. Ion exchange resins require concentrated solutions of acid, base, or salt for regeneration, whereas carbon aerogel electrodes require only electrical discharge or reverse polarization. Aqueous solutions of NaCl, NaNO <sub>3</sub> , NH <sub>4</sub> ClO <sub>4</sub> , Na <sub>2</sub> CO <sub>3</sub> , Na <sub>2</sub> SO <sub>4</sub> , and Na <sub>3</sub> PO <sub>4</sub> have been separated into concentrate and high-purity product streams. The deionization of NaCl solution with two parallel stacks of carbon aerogel electrodes in a potential-swing mode is discussed in detail. The selective removal of Cu, Zn, Cd, Pb, Cr, Mn, Co, and U from a variety of process solutions and natural waters has also been demonstrated. Feasibility tests indicate that the remediation of Cr(VI)-contaminated ground water may be possible.				
<b>14. SUBJECT TERMS</b>  Carbon aerogel electrodes, deionization of water, heavy metal removal from water, SERDP			<b>15. NUMBER OF PAGES</b> 11	
			<b>16. PRICE CODE</b> N/A	
<b>17. SECURITY CLASSIFICATION OF REPORT</b>  unclass.	<b>18. SECURITY CLASSIFICATION OF THIS PAGE</b>  unclass.	<b>19. SECURITY CLASSIFICATION OF ABSTRACT</b>  unclass.	<b>20. LIMITATION OF ABSTRACT</b> UL	

19980709 159

## **The Use of Carbon Aerogel Electrodes for Deionizing Water and Treating Aqueous Process Wastes**

Joseph C. Farmer, Gregory V. Mack and David V. Fix  
Lawrence Livermore National Laboratory  
Livermore, California 94550

### **Abstract**

A wide variety of ionic contaminants can be removed from aqueous solutions by electrosorption on carbon aerogel electrodes. Carbon aerogel is an ideal electrode material because of its low electrical resistivity ( $\leq 40 \text{ m}\Omega\text{-cm}$ ), high specific surface area (400 to 1100  $\text{m}^2/\text{g}$ ), and controllable pore size distribution ( $\leq 50 \text{ nm}$ ). This approach may avoid the generation of a substantial amount of secondary waste associated with ion exchange processing. Ion exchange resins require concentrated solutions of acid, base, or salt for regeneration, whereas carbon aerogel electrodes require only electrical discharge or reverse polarization. Aqueous solutions of NaCl, NaNO<sub>3</sub>, NH<sub>4</sub>ClO<sub>4</sub>, Na<sub>2</sub>CO<sub>3</sub>, Na<sub>2</sub>SO<sub>4</sub> and Na<sub>3</sub>PO<sub>4</sub> have been separated into concentrate and high-purity product streams. The deionization of a 100  $\mu\text{S/cm}$  NaCl solution with two parallel stacks of carbon aerogel electrodes in a potential-swing mode is discussed in detail. The selective removal of Cu, Zn, Cd, Pb, Cr, Mn, Co and U from a variety of process solutions and natural waters has also been demonstrated. Feasibility tests indicate that the remediation of Cr(VI)-contaminated ground water may be possible.

### **Introduction**

Lawrence Livermore National Laboratory (LLNL) has developed an electrically-regenerated electrosorptive separation process for removing ionic impurities from aqueous waste streams as a minimally-polluting, energy-efficient, and potentially cost-effective alternative to ion exchange, reverse osmosis, electrodialysis, and evaporation. Aqueous solutions with various anions and cations are passed through a stack of carbon aerogel electrodes, each having a very high specific surface area (400 to 1100  $\text{m}^2/\text{g}$ ) and exceptionally low electrical resistivity ( $\leq 40 \text{ m}\Omega\text{-cm}$ ). After polarization, ions are removed from the electrolyte by the imposed electric field and electrosorbed on electrode surfaces. For example, aqueous solutions of NaCl, NaNO<sub>3</sub>, NH<sub>4</sub>ClO<sub>4</sub>, Na<sub>2</sub>CO<sub>3</sub>, Na<sub>2</sub>SO<sub>4</sub>, and Na<sub>3</sub>PO<sub>4</sub> have been separated into two streams, one being relatively pure water and the other a relatively concentrated salt solution. Trace quantities of various heavy metals such as Cu, Ni, Pb, Co, Cr, Mn and U have also been removed from process solutions and natural waters by electrosorption.

### **Background**

Ion exchange is used for deionization of process water and to remove heavy metals from aqueous waste streams, as well as contaminated ground water. Unfortunately, the chemical regeneration of saturated ion exchange resin produces a significant amount of secondary waste. A variety of electrochemical alternatives to ion exchange are now under investigation. For example, polypyrrole films on reticulated vitreous carbon electrodes are being used for in situ reduction of

Cr(VI) [1]. This approach appears to suffer from a gradual degradation of the electrodes due to a loss of polypyrrole. Another process involves the use of electrodes coated with films of electroactive ferrocyanide for selective removal  $\text{Cs}^+$  from solutions of sodium salts [2]. Here too electrode life is limited by the stability of the electroactive film. Carbon aerogel electrodes appear to have the necessary selectivity and stability to enable them to serve as a viable alternative to ion exchange.

Several publications have appeared that discuss the use of porous electrodes for the recovery of heavy metals from aqueous solutions by electrodeposition [3-6]. The first studies of flow-through capacitors with activated carbon electrodes applied to desalination appear to have been published at the University of Oklahoma in the early 1960's [7,8]. Johnson et al. conducted similar studies of reversible electrosorption with beds of activated carbon in the early 1970's [9-11]. Johnson's work prompted Newman to develop a comprehensive theoretical model for the capacitive charging of porous carbon electrodes [12]. Additional work was done in Israel several years later and published in the 1980's [13-15]. Several practical problems are encountered with conventional activated carbon systems. Electrode performance has been found to degrade with time. Furthermore, polymeric binders occlude significant fractions of the activated carbon surface and are susceptible to radiation damage. Electrochemical cells that use flow-through beds of activated carbon powder as electrodes require membrane separators for electrical insulation and to prevent entrainment of individual particles in the flow. Flow through such porous media is characterized by high pressure drop. Finally, process efficiency is lowered by the large potential drop that develops in thick electrodes and packed beds.

Carbon aerogel is an ideal electrode material because of its low electrical resistivity ( $\leq 40 \text{ m}\Omega\text{-cm}$ ), high specific surface area (400 to  $1100 \text{ m}^2/\text{g}$ ), and controllable pore size distribution ( $\leq 50 \text{ nm}$ ). Resorcinol-formaldehyde (RF) aerogels and their carbonized derivatives, including CAC, were developed by LLNL [16-19]. Note that thermal and chemical activation has been used to achieve specific surface areas of  $2600 \text{ m}^2/\text{g}$  [20]. The exceptionally high conductivity of carbon aerogel, in contrast to loosely bonded carbon powders or activated carbon fiber cloths (ACFCs), is attributable to its monolithic structure which is composed of interconnected, covalently-bonded carbon particles ( $\sim 12 \text{ nm}$  diameter). In contrast to electrodes made from activated carbon powders and fibers, the activation energy for carrier transport in carbon aerogel is relatively small. As a result of these desirable characteristics, carbon aerogel has already been used as electrodes in a supercapacitor with high energy and power density [21]. However, this device was not designed to permit electrolyte flow and requires membranes to physically separate the electrodes. More recently, carbon aerogel electrodes developed by LLNL have been employed in a variety of configurations to remove ionic contaminants from water [22-31].

## **Experimental**

Double-sided electrodes for the electrochemical cell were made by gluing two sheets of carbon aerogel composite (CAC) to both sides of a titanium plate with graphite-filled epoxy. The titanium plate serves as both a current collector and a structural support. Each sheet of CAC was  $6.9 \text{ cm} \times 6.9 \text{ cm} \times 0.0127 \text{ cm}$  and had a total active surface of approximately  $2.8 \times 10^6 \text{ cm}^2$ . A stack of 192 double-sided, titanium-supported electrodes had a total active surface area of

approximately  $1.1 \times 10^9 \text{ cm}^2$ . Electrolyte flowed through the stack in open channels formed between adjacent electrodes. An electrode separation of 0.12 to 0.16 cm was maintained by plastic spacers and rubber compression seals. By arranging the electrodes so that orifices alternate from one side of the stack to the other, flow from the bottom of the stack to the top is serpentine.

It is noteworthy that activated carbon powders with Brunauer-Emmet-Teller (BET) surface areas as high as  $3000 \text{ m}^2/\text{g}$  are readily available. However, much of the surface area in such materials is located inside pores having diameters less than 1 nm. It is believed that the electrochemically active area is only a fraction of the BET surface area. BET analyses are probably misleading since gas molecules can penetrate much smaller pores than a typical electrolyte. For example, the bond length of  $\text{N}_2$  is only 0.1 nm. It is very doubtful that this level of porosity contributes to electrochemical double layer formation since electrolyte penetration and double layer formation are questionable on this scale. From the Gouy-Chapman theory, as well as the Stern modification of that theory, it is believed that a fully-developed electric double layer on a planar electrode with no detrimental shielding effects would require much greater distances for full development. In the case of a 1:1 electrolyte in water at  $25^\circ\text{C}$ , the characteristic thickness of the diffuse layer ranges from 1 nm at a concentration of  $10^{-1} \text{ M}$  to 30 nm at  $10^{-4} \text{ M}$  [32,33].

A potential-swing electrosorption process based on carbon aerogel electrodes has been developed and demonstrated. This prototype can produce a continuous flow of both product water and concentrate by operating two stacks of electrodes in parallel. One stack purifies while the other is electrically regenerated. Such synchronous operation requires a significant degree of automation and sophistication, which has been incorporated into the demonstration process now available at LLNL [31]. Flow is generated by a programmable, magnetically-coupled, screw pump with a 304 stainless steel head. All lines are made of Teflon and had a nominal diameter of 0.635 cm (1/4 inch). The cells are polarized by programmable power supplies that have a voltage range of 0 to 12 V or a current range of 0 to 60 A. Sensors were placed on the inlet and outlet lines of the electrode stack. Electrical conductivity of the solution, pH, individual ion concentrations, and temperature are continuously monitored. The system is controlled by a personal computer. A single AT-MIO-16DH data acquisition board, installed in the computer, provides the interface to the Input-Output (I/O) Signal Subsystem. The I/O subsystem consists of a single 12-slot Signal Conditioning Extension Interface (SCXI) chassis. The chassis contains seven 8-channel analog-to-digital (A/D) modules to measure flow, level, pressure, temperature, pH, and conductivity; two 16-channel single-pole double-throw (SPDT) relay modules for controlling pumps and valves; and one 6-channel digital-to-analog (D/A) module for controlling power supply voltages and pump speed. The SCXI hardware provides multiplexing, filtering, isolation, and amplification for the process signals. The operating system is DOS v6.22 running Windows v3.1. LabVIEW v3.1 software, running under Windows, is used for data acquisition and control. LabVIEW is a graphical programming environment which provides integrated tools for acquisition, control, analysis, and presentation, as well as connectivity to serial, parallel, voltage, current loop, RTD, thermistor, and relay communication interfaces. Data acquisition and control software is optimized and converted to compiled run-time code. Operator input is via a mouse and keyboard.

Studies of the removal of chromium from ground water employed a stack of 192 electrodes similar to the one previously described. Experiments with heavy metals in 0.1 M  $\text{KNO}_3$  electrolyte were conducted with two single-sided, titanium-supported electrodes in an undivided cell. A 500 ml volume of electrolyte was continuously recycled at a rate of 50 ml/min. Additional experiments were conducted with a divided cell that employed an ion exchange membrane as an electrode separator. The membrane facilitated regeneration by voltage reversal. Experiments with heavy metals in sea water were conducted with 40 sheets of unsupported CAC separated by a polypropylene net (InterNet Incorporated, Product No. XN-1678, 0.125 inch mesh) in an undivided, plastic cell. A 1000 ml volume of sea water was continuously recycled at a rate of 50 ml/min. In experiments involving heavy metals, samples of both the influent and effluent waters were withdrawn and analyzed for various constituents at LLNL and outside laboratories. Trace quantities of heavy metals such as Cu, Ni, Co, Pb, Mn, Cr and U were determined with inductively coupled plasma with mass spectrometry (ICP-MS). Total chromium and Cr(VI) were measured by standard methods [34-36]. Atomic absorption spectroscopy was used to measure the total concentration of chromium as specified by EPA 218.2. Aqueous spectrophotometry was used to measure the concentration of Cr(VI) alone as specified by EPA 7196. The detection limit of this standard method for Cr(VI) was lowered from 5 to 2 ppb by the use of a spectrophotometer cell with a 5 cm optical path.

## Results

The deionization of a 100  $\mu\text{S}/\text{cm}$  NaCl solution with two parallel stacks in a potential-swing mode is illustrated by Figure 1. Each stack had 192 double-sided, titanium-supported electrodes. In this early experiment, the flow rate was 25 ml/min during both deionization and regeneration. The electrodes were polarized at 1.2 V during deionization and discharged at 0 V during regeneration. A continuous stream of product with an average conductivity of 10 to 15  $\mu\text{S}/\text{cm}$  was generated. Peaks in the conductivity of the concentrate stream were coincident with regeneration steps for stacks A and B, respectively. From the differences in peak height, it is evident that stack A had less electrosorption capacity than stack B. The difference is attributed to a change in electrode fabrication. These data illustrate the importance of using balanced stacks to minimize "waiting" and to maximize throughput. The effects of concentration and voltage are discussed in earlier publications [26,27].

Trace quantities of various heavy metals such as Cu, Zn, Cd, Pb, Cr, Mn, Co and U were removed from process solutions and natural waters by electrosorption on CAC electrodes. As shown in Figure 2, two single-sided, titanium-supported electrodes were used to remove Cu, Zn, Cd, and Pb from 500 ml of 0.1 M  $\text{KNO}_3$  with electrosorption capacities ranging from  $5 \times 10^{-4}$  to  $9 \times 10^{-4}$  gram of heavy metal per gram of CAC (g/g-CAC). As shown in Figure 3, forty unsupported CAC sheets were used as electrodes to remove trace quantities of Co, Cr, Mn, Pb, and U from 1000 ml of sea water. In this case, slight corrosion of auxiliary equipment appears to have released detectable levels of Ni (not shown). The polarization was 1.2 V and the circulation rate was 50 ml/min in both Figures 2 and 3.

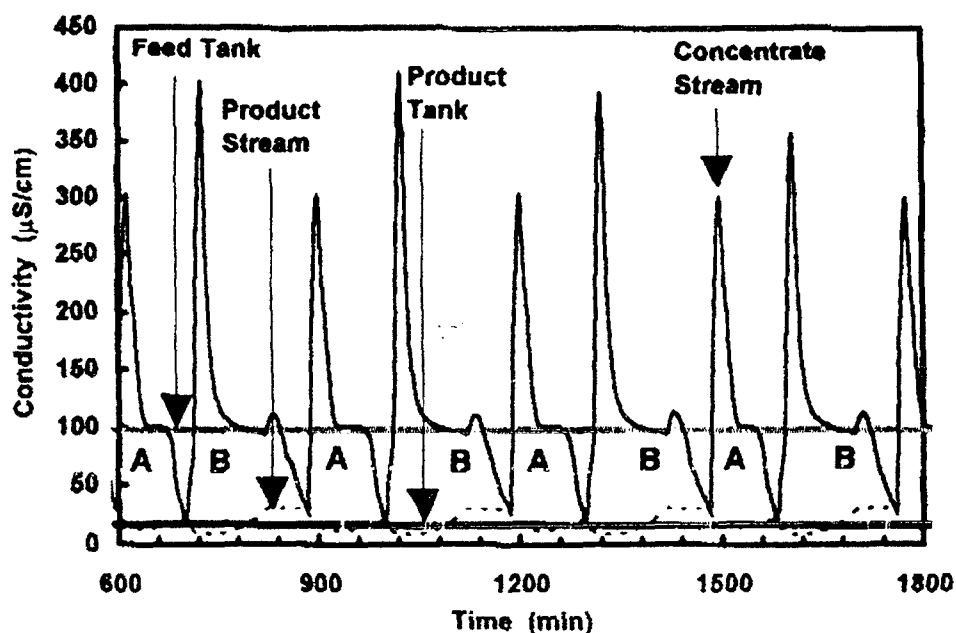


Figure 1. Deionization of 100  $\mu\text{S/cm}$  NaCl solution in potential-swing mode.

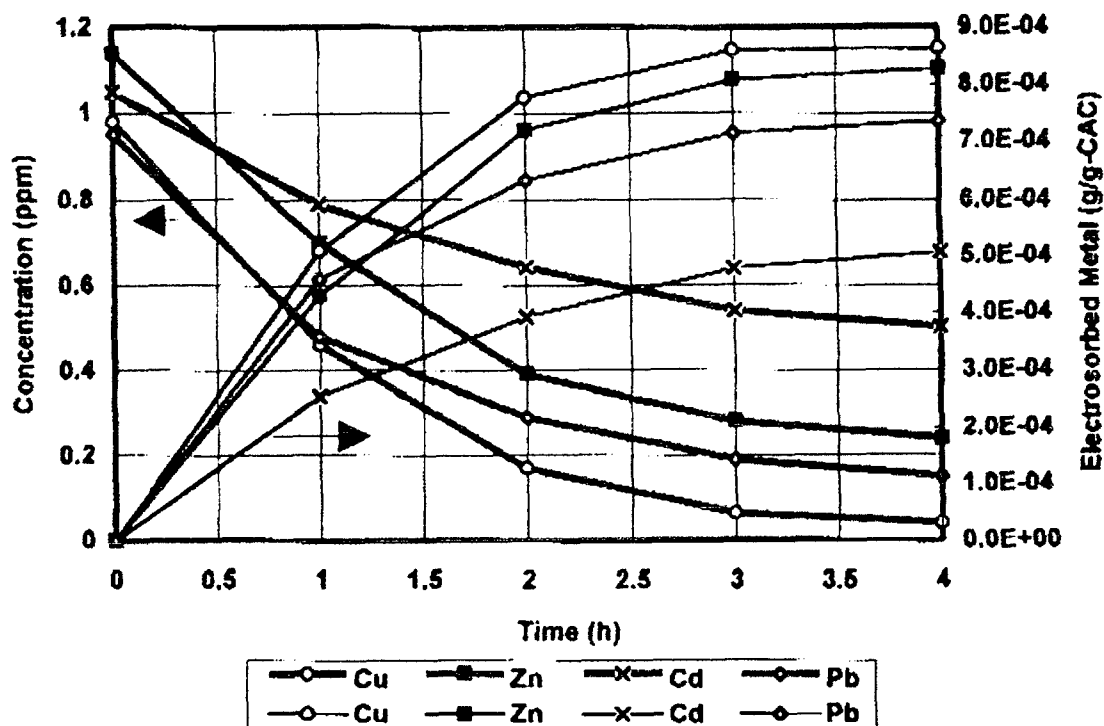


Figure 2. Two carbon aerogel electrodes polarized at 1.2 V in an undivided cell. Removal of 1 ppm heavy metal contamination from a 500 ml of 0.1 M  $\text{KNO}_3$  with complete recycle at 50 ml/min.

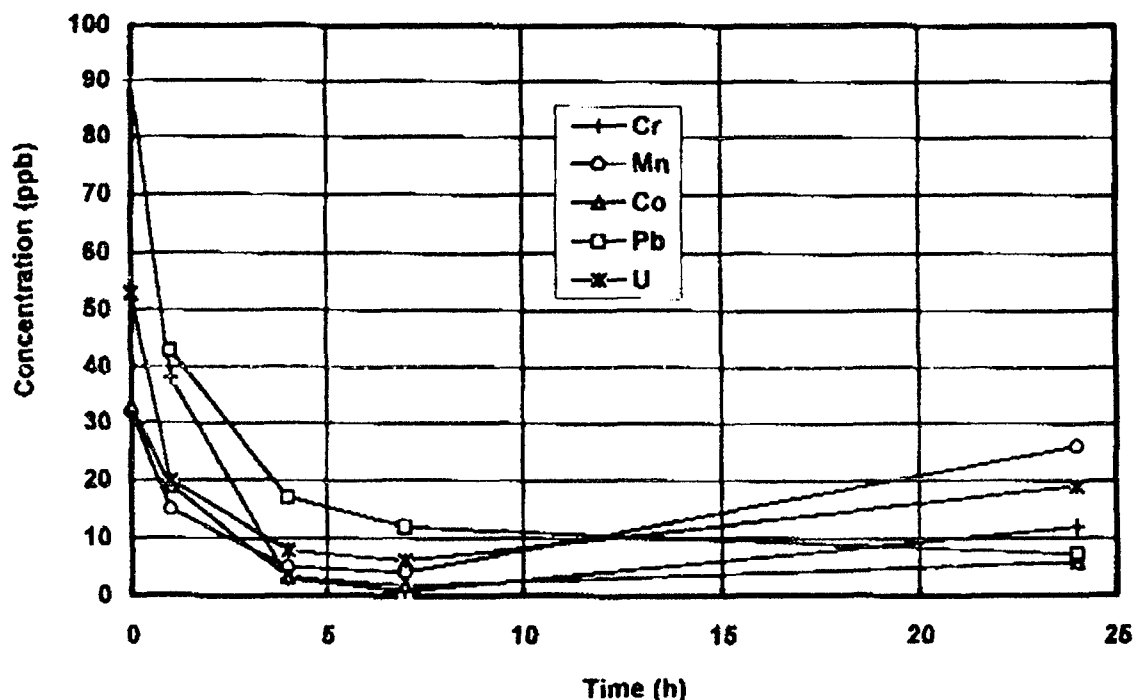


Figure 3. Removal of heavy metals at initial concentrations of ~100 ppb from 1000 ml of sea water with forty unsupported CAC sheets polarized at 1.2 V. Electrode separation in each undivided cell was maintained by polyethylene filtration netting.

Laboratory-based treatability studies at LLNL have shown that chromium can be selectively removed from contaminated ground water with a 530 ppm TDS background. A stack of 192 double-sided, titanium-supported electrodes was used in the laboratory for the continuous removal of chromium from a 100 ml/min stream of contaminated ground water with 530 ppm total dissolved solids (TDS). As illustrated by Figure 4, the total concentration of chromium was lowered from 32 to 2 ppb, well below the acceptable Cr(VI) level of 11 ppb for surface water discharge. Essentially all of the chromium in the inlet stream was Cr(VI), which was probably in the form of  $\text{HCrO}_4^-$ ,  $\text{CrO}_4^{2-}$ , or  $\text{Cr}_2\text{O}_7^{2-}$ . Any Cr(III) present was probably in the form of  $\text{Cr}(\text{OH})^{2+}$ ,  $\text{Cr}(\text{OH})_2^+$ ,  $\text{Cr}(\text{OH})_4^+$ , or  $\text{CrO}_2^-$  [36]. The possibility of forming non-ionic chromium hydroxide monomer, dimer, trimer, and tetramer is also recognized. Carbon aerogel electrodes have also been used to remove Cr(VI) from ground water during a field test [37]. The total concentration of chromium in the outlet stream was lowered from 30-35 ppb to less than 7 ppb with a stack of 144 electrodes polarized at 1.2 V. The speciation of chromium in the outlet stream was approximately 30% Cr(III) and 70% Cr(VI). It is evident that in addition to removing Cr(VI) from the stream of water, Cr(VI) was reduced to Cr(III). Furthermore, the chromium removal continued long after saturation of the carbon aerogel with other ionic species, indicating a high degree of selectivity towards chromium. Most of the chromium was recovered after

regeneration by (1) discharge at 0 V and 100 ml/min, (2) reverse polarization at -1.2 V and 1000 ml/min, and (3) discharge at 0 V and 1000 ml/min. In this case, regeneration was achieved in an undivided cell.

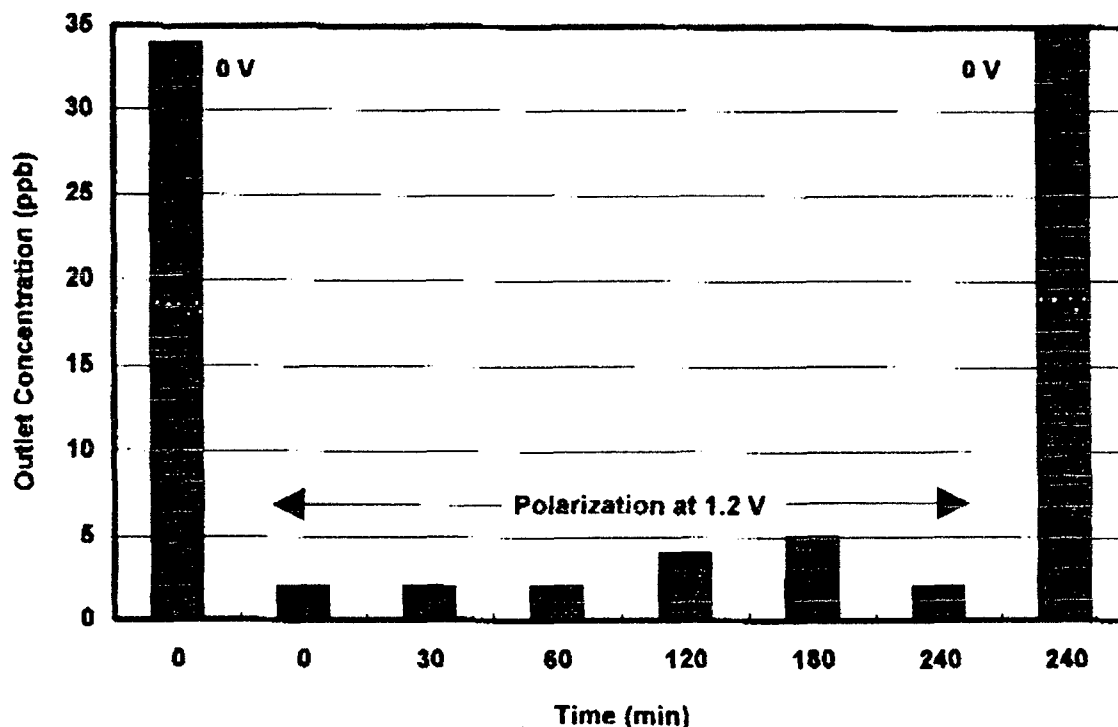


Figure 4. Selective removal of 35 ppb chromium from ground water with 530 ppm TDS during initial laboratory test.

### Summary

This reversible aerogel-based electrosorption process avoids the secondary waste associated with the chemical regeneration of ion exchange columns. Unlike ion exchange, no acids, bases, or salt solutions are required for regeneration of the system. By using reversible electrosorption as an alternative to ion exchange, it may be possible to eliminate approximately 100 kg of spent chemical regenerant per kg of regenerated resin. Regeneration is accomplished by electrical discharge or reverse polarization. The carbon aerogel process can produce a continuous flow of product water by operating two stacks of carbon aerogel electrodes in parallel (potential-swing mode). One stack purifies while the other is electrically regenerated. In cases where separation is due to double layer charging, energy recovery may be possible by using the energy released during the discharge of one stack (regeneration) to charge the other (deionization). This generic process design should facilitate energy recovery in industrial-scale systems. An industrial grade, computerized control system with a graphical user interface has been fully developed to synchronize the operation of parallel stacks of carbon aerogel electrodes. Electrosorption processes based on carbon aerogel promise to be much more energy efficient than comparable



thermal processes and appear to have operational advantages over electrodialysis and reverse osmosis since membranes and high-pressure pumps are not necessarily required [38]. Work is now being done to increase the capacity of the carbon aerogel electrosorption process to practical levels. It has been demonstrated that a large quantity of high-quality CAC sheets (4000 ft<sup>2</sup>) can be mass produced at an apparent cost of approximately \$50 per square (GenCorp-Aerojet Plant in Sacramento, California). It may be possible to reduce the cost to less than \$2 per square foot in the future.

### **Acknowledgment**

The outstanding analytical work by A. M. Volpe, R. E. Martinelli, A. K. Vu, and K. L. Carrol is gratefully acknowledged. Contributions of R. W. Pekala in preparation of carbon aerogel is also acknowledged. Funding for this project was provided by the Strategic Environmental Research and Development (SERDP) Program. This work was done under the auspices of the U.S. Department of Energy (DOE) by Lawrence Livermore National Laboratory (LLNL) under Contract No. W-7405-Eng-48.

### **References**

1. L. F. Fernandez, J. G. Ibanez, K. Rajeshwar, S. Basak, "The Reduction of Cr(VI) with Polypyrrole on Reticulated Vitreous Carbon: Miscellaneous Effects," Abs. No. 524, 189th Mtg. Electrochem. Soc., Los Angeles, CA, May 5-10, 1996, Electrochem. Soc., Pennington, NJ, Ext. Abs., Vol. 96-1, p. 691 (1996).
2. M. Lilga, J. Sukanto, M. Buehler, "Electrically Controlled Cesium Ion Exchange," Proc. 1996 Annual Technical Exchange Meeting, Efficient Separations and Processing Crosscutting Program, Gathersburg, MD, Jan. 16-19, 1996, pp. 47-52 (1996)
3. J. A. Trainham, J. Newman, "Flow-Through Porous Electrode Model: Application to Metal-Ion Removal from Dilute Streams," J. Electrochem. Soc., Vol. 124, No. 10, pp. 1528-1540 (1977).
4. Van Zee, J. Newman, "Electrochemical Removal of Silver Ions from Photographic Fixing Solutions Using a Porous Flow-Through Electrode," J. Electrochem. Soc., Vol. 124, No. 5, pp. 706-708 (1977).
5. M. Matlosz, J. Newman, "Experimental Investigation of a Porous Carbon Electrode for the Removal of Mercury from Contaminated Brine," J. Electrochem. Soc., Vol. 133, No. 9, pp. 1850-1859 (1986).
6. D. Pletcher, "Industrial Electrochemistry," Chapter 11, Water Treatment and Environmental Protection, Chapman and Hall, London, UK, p. 291 (1982).
7. B. B. Arnold, G. W. Murphy, "Studies on the Electrochemistry of Carbon and Chemically Modified Carbon Surfaces," J. Phys. Chem., Vol. 65, No. 1, pp. 135-138 (1961).

8. D. D. Caudle, J. H. Tucker, J. L. Cooper, B. B. Arnold, A. Papastamatakis, "Electrochemical Demineralization of Water with Carbon Electrodes," Research and Development Progress Report No. 188, United States Department of Interior, 190 pages, May (1966).
9. A. M. Johnson, A. W. Venolia, J. Newman, R. G. Wilbourne, C. M. Wong, W. S. Gillam, S. Johnson, R. H. Horowitz, "Electrosorb Process for Desalting Water," Office of Saline Water Research and Development Progress Report No. 516, U.S. Dept. Interior Pub. 200 056, 31 pages, March (1970).
10. A. M. Johnson, A. W. Venolia, R. G. Wilbourne, J. Newman, "The Electrosorb Process for Desalting Water," Marquardt Co., Van Nuys, CA, 36 pages, March (1970).
11. A. M. Johnson, "Electric Demineralizing Apparatus," U.S. Pat. No. 3515664, June 2 (1970).
12. A. M. Johnson, J. Newman, "Desalting by Means of Porous Carbon Electrodes," J. Electrochem. Soc., Vol. 118, No. 3, pp. 510-517 (1971).
13. Y. Oren, A. Soffer, "Electrochemical Parametric Pumping," J. Electrochem. Soc., Vol. 125, No. 6, pp. 869-875 (1978).
14. Y. Oren, A. Soffer, "Water Desalting by Means of Electrochemical Parametric Pumping: I. The Equilibrium Properties of a Batch Unit Cell," J. Appl. Electrochem., Vol. 13, No. 4, pp. 473-487 (1983).
15. Y. Oren, A. Soffer, "Water Desalting by Means of Electrochemical Parametric Pumping: II. Separation Properties of a Multistage Column," J. Appl. Electrochem., Vol. 13, No. 4, pp. 489-505 (1983).
16. R. W. Pekala, S. T. Mayer, J. F. Poco, J. L. Kaschmitter, "Structure and Performance of Carbon Aerogel Electrodes," in Novel Forms of Carbon II, C. L. Renschler, D. M. Cox, J. J. Pouch, Y. Achiba, Eds., MRS Symp. Proc., Vol. 349, No. 79 (1994).
17. J. Wang et al., "Carbon Aerogel Composite Electrodes," Anal. Chem., Vol. 65, pp. 2300-2303 (1993).
18. R. W. Pekala, C. T. Alviso, in Novel Forms of Carbon, C. L. Renschler, J. J. Pouch, D. M. Cox, Eds., MRS Symp. Proc., Vol. 270, No. 3 (1992).
19. R. W. Pekala, in Ultrastructure Processing of Advanced Materials, D. R. Uhlmanjn, D. R. Ulrich, Eds., John Wiley and Sons, Inc., New York, NY, pp. 711-717 (1992).
20. Y. Hanzawa, K. Kaneko, R. W. Pekala, M. S. Dresselhaus, "Activated Carbon Aerogels," unpublished manuscript (1996).

21. S. T. Mayer, R. W. Pekala, J. L. Kaschmitter, "The Aerocapacitor: An Electrochemical Double-layer Energy-Storage Device," *J. Electrochemical Society*, Vol. 140, No. 2, pp. 446-451 (1993).
22. J. C. Farmer, "Method and Apparatus for Capacitive Deionization, Electrochemical Purification, and Regeneration of Electrodes," U.S. Pat. No. 5428858, June 20 (1995).
23. J. C. Farmer, D. V. Fix, G. V. Mack, R. W. Pekala, J. F. Poco, "The Use of Capacitive Deionization with Carbon Aerogel Electrodes to Remove Inorganic Contaminants from Water," *Proc. 1995 Intl. Low-Level Conf.*, Orlando, Florida, July 10-12, 1995, Electric Power Research Institute, Palo Alto, CA, Rept. TR-105569, pp. 42 1-23 (1995).
24. J. C. Farmer, D. V. Fix, G. V. Mack, R. W. Pekala, J. F. Poco, "Capacitive Deionization of Water: An Innovative New Process," *Proc. Fifth Intl. Conf. Rad. Waste Mgmt. Environ. Remediation*, Berlin, Germany, September 3-9, 1995, American Society of Mechanical Engineers (ASME), New York, NY, Vol. 2, pp. 1215-1220 (1995).
25. J. C. Farmer, D. V. Fix, G. V. Mack, R. W. Pekala, J. F. Poco, "Capacitive Deionization with Carbon Aerogel Electrodes: Carbonate, Sulfate, and Phosphate," *Proc. 1995 Intl. SAMPE Tech. Conf.*, Albuquerque, NM, October 9-12, 1995, Society for the Advancement of Material and Process Engineering (SAMPE), Covina, CA, Vol. 27, pp. 294-304 (1995).
26. J. C. Farmer, D. V. Fix, G. V. Mack, R. W. Pekala, J. F. Poco, "Capacitive Deionization of NaCl and NaNO<sub>3</sub> Solutions with Carbon Aerogel Electrodes," *J. Electrochem. Soc.*, Vol. 143, No. 1, pp. 159-169 (1996).
27. J. C. Farmer, D. V. Fix, G. V. Mack, R. W. Pekala, J. F. Poco, "Capacitive Deionization of NH<sub>4</sub>ClO<sub>4</sub> Solutions with Carbon Aerogel Electrodes," *J. Appl. Electrochem.*, Vol. 26 (Ms. No. NA-025-95, in press) (1996).
28. J. C. Farmer, D. V. Fix, R. W. Pekala, J. K. Nielsen, A. M. Volpe, D. D. Dietrich, "The Use of Carbon Aerogel Electrodes for Environmental Cleanup," *Proc. Symposium on the Production and Use of Carbon-Based Materials for Environmental Cleanup*, 1996 Spring Meeting of the American Chemical Society, New Orleans, Louisiana, March 24-29, 1996, Vol. 41, p. 484 (1996).
29. J. C. Farmer, R. W. Pekala, F. T. Wang, D. V. Fix, A. M. Volpe, D. D. Dietrich, W. H. Siegel, "Electrochemical and Photochemical Treatment of Aqueous Waste Streams," *Proc. Spectrum 96*, Nucl. Haz. Waste Mgmt. Intl. Topical, , Seattle, Washington, August 18-23, 1996, Am. Nucl. Soc., La Grange Park, IL (1996).
30. J. C. Farmer, S. M. Bahowick, J. E. Harrar, D. V. Fix, R. E. Martinelli, A. K. Vu, K. L. Carroll, "Electrosorption of Chromium Ions on Carbon Aerogel Electrodes as a Means of Remediating Ground Water," *U. Calif., Lawrence Livermore Natl. Lab. Rept.* (1996).

31. G. V. Mack, J. C. Farmer, D. V. Fix, G. W. Johnson, D. W. O'Brien, "Design of the Control System for the Continuous-Flow Potential-Swing Capacitive Deionization Process," U. Calif., Lawrence Livermore Natl. Lab. Rept. No. UCRL-ID-117626, September 30 (1994).
32. A. J. Bard, L. R. Faulkner, Electrochemical Methods, Fundamentals and Applications, John Wiley and Sons, New York, NY, pp. 500-515 (1980).
33. J. S. Newman, Electrochemical Systems, 2nd Ed., Prentice Hall, Englewood Cliffs, NJ, pp. 157-185 (1991).
34. U. S. Environmental Protection Agency, "Test Methods for Evaluating of Solid Waste," SW-846, 3rd Ed., Vol. 1A, Ch. 3.3 (1986).
35. U. S. Environmental Protection Agency, "Methods for Chemical Analysis of Water and wastes," Report EPA 600/4-79-020 (1983).
36. Code of Federal Regulations, Title 40, Part 36, Appendix A (1993).
37. F. Y. Saleh, G. E. Mbamalu, Q. H. Jaradat, C. E. Brungardt, "Ion Chromatography - Photodiode Array UV-Visible Detection of Cr(III) Hydrolytic Polymerization Products in Pure and Natural Waters," *Analytical Chem.*, Vol. 68, No. 5, pp. 740-745 (1996).
38. C. J. King, Separation Processes, 2nd Ed., McGraw-Hill, San Francisco, CA, pp. 661-664 (1980).